1965

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http://hdl.handle.net/10945/12274

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Submitted in partial fulfillment of
the requirements for the degree of

MASTER OF SCIENCE
IN
PHYSICS

United States Naval Postgraduate School
Monterey, California

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ABSTRACT

Half-lives of thermal neutron activated isomers were determined by direct decay measurements with a scintillation counter. Scaler pulses following a single channel analyzer, together with precise timing pulses, were traced concurrently by a fast chart recorder. The gain stability of the detecting circuit was also monitored during the decay period. Decay counts were corrected for counting circuit dead-time and for pulse pile-up, and analyzed by a digital computer. The average half-lives obtained, together with their statistical uncertainties are:

Hf^{178m} 3.79 ± 0.07 sec.
Hf^{179m} 18.18 ± 0.28 sec.
Yb^{177m} 6.40 ± 0.02 sec.
W^{183m} 5.14 ± 0.02 sec.

The authors wish to express their appreciation to Mr. H. L. McFarland for his assistance in operating the nuclear reactor, and to Mrs. Mary Barker and Mrs. Thelma Tegtmeier for their assistance in punching numerous computer data cards. The authors also acknowledge the preliminary studies made in this field by Griggs and Macdonald [10] and the invaluable assistance and guidance of Prof. G. W. Rodeback in all phases of the investigation.

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1. Introduction

The objective of this investigation has been to determine, as accurately as possible, the half-lives of the neutron induced isomers, Hf^{178m}, Hf^{179m}, $Yb^{177m'}$, and W^{183m}. All of their half-lives are less than twenty seconds. In theory, a half-life measurement is a relatively straight-forward and uncomplicated procedure. The decaying isomer is observed for several half-lives so that a decay rate versus time relationship is determined. Utilizing the fact that radioactive decay is an exponential function, this experimentally determined decay rate is analyzed mathematically or graphically to determine the desired decay constant or half-life.

In practice, when high accuracy is desired, the procedure is complicated by several factors, any one of which may cause significant errors in the determination. The first of these complicating factors is the fact that not all of the gamma photons that produce a pulse in the scintillation counter are recorded as separate pulses by the counting system. Inherent in any electronic counting system is a characteristic dead-time, during which decays are not recorded. Also, difficulty in counting results from the phenomenon of pile-up in which pulses from two separate decays, occuring simultaneously or nearly so, add by superposition and result in one pulse whose amplitude is the sum of the amplitudes of the two individual pulses. Another major source of difficulty lies in the occurence of gain shift, which occurs in a photomultiplier tube when it is subjected to

changes in counting rate, and which may be particularly important in the study of decays with half-lives of the order of a few seconds. The result of such a shift in gain is an observed count rate that is either too high or too low. Other possible sources of error may arise from the presence of contaminating radio-nuclides, and from instabilities in the electronic circuitry which defines the pulse height interval over which pulses are recorded.

In this investigation the gamma radiation associated with the isomeric transitions was detected with a scintillation counter. Detected transitions were recorded using a linear high-overload amplifier and analyzer, a high speed scaler, and a fast chart recorder. The count rates so determined were corrected for pile-up and dead-time losses by applying experimentally and mathematically determined correction factors. A monitor system was utilized whereby the gain stability of the detector could be measured and suitable corrections applied, if needed. Counting data, properly corrected, were then analyzed by an electronic computer to determine the half-lives under study. Where it has been found impossible to observe an isomer singly, due to the presence of other nuclides, the overall decay curve has been mathematically analyzed, allowing the curve to be decomposed into its several components.

2. Experimental Equipment (Schematic, Figure 2-1).

Neutron irradiated samples of the isomers were transferred from the AGN-201 reactor* to the scintillation counter by means of a high speed pneumatic transfer system. Pulses from the counter were fed through two amplifier channels, one analyzing pulses from the isotope being examined and the other analyzing pulses from a "gain-monitoring" alpha source. Pulses were fed by the respective amplifiers into high speed scalers, which in turn fed counting information into continuous-flow strip type chart recorders. Standardized time pulses were simultaneously fed into each of these chart recorders. See Figure 2-2.

a. Transfer system

The high-speed pneumatic transfer system for use with the AGN-201 reactor was built at USNPGS by Paas and Sullivan [7]. Samples to be radiated were semi-permanently enclosed in gelatin capsules which were further encased in balsa wood projectiles. The projectile containing the sample which was to be radiated was placed in a pneumatic tube in which a vacuum-positive pressure differential could be applied. This pressure differential moves the sample rapidly and seats it in the core of the reactor where it is irradiated by thermal neutrons. After the desired irradiation period, the direction of pressure differential is rapidly reversed, moving the "hot" sample from the reactor core into counting position above the

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* The AGN-201 Reactor is a graphite and polyethylene moderated research reactor capable of producing a neutron flux up to 4 x 10^{10} n/cm^2-sec at the AEC license limit of 1000 watts.

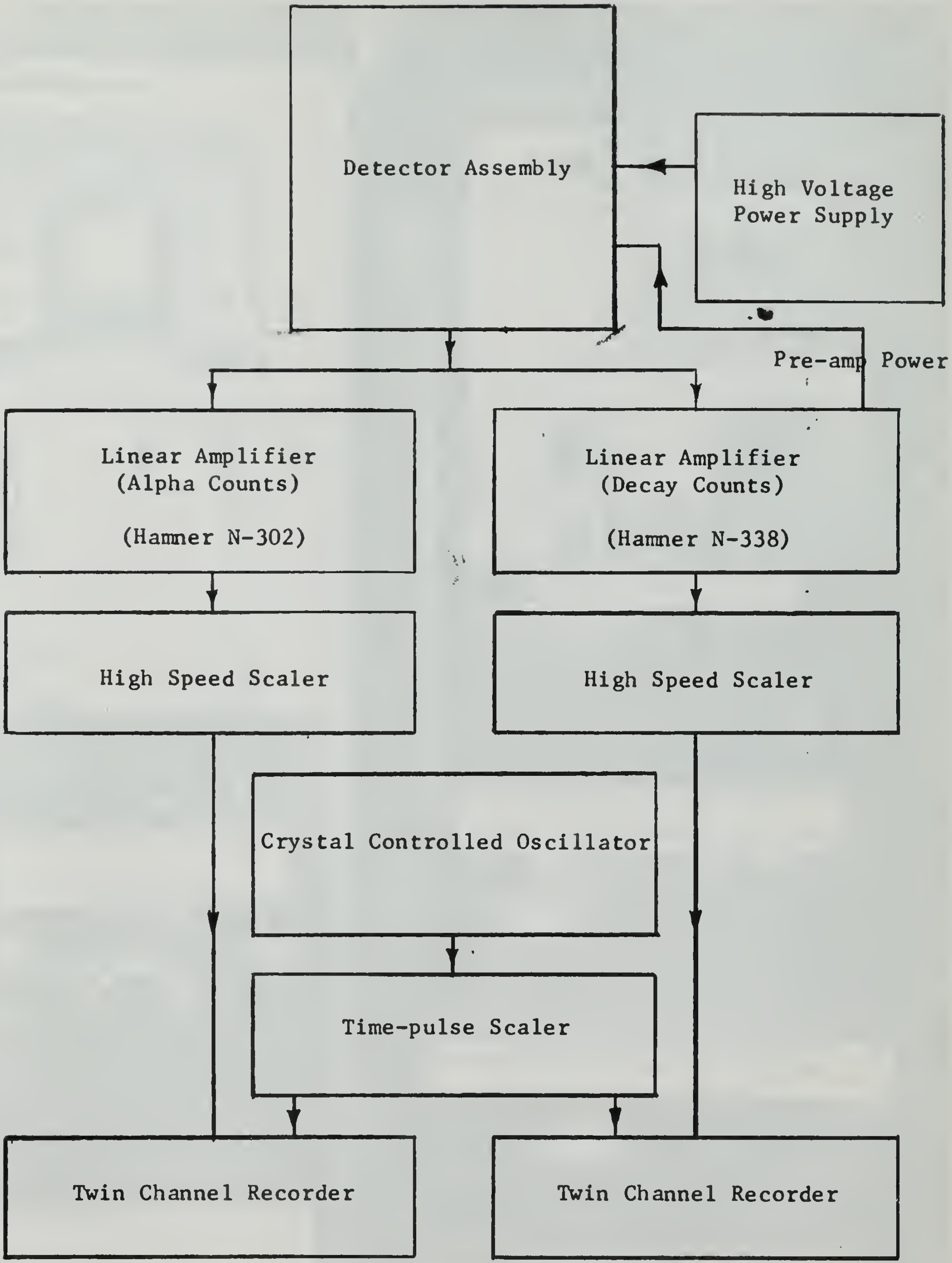
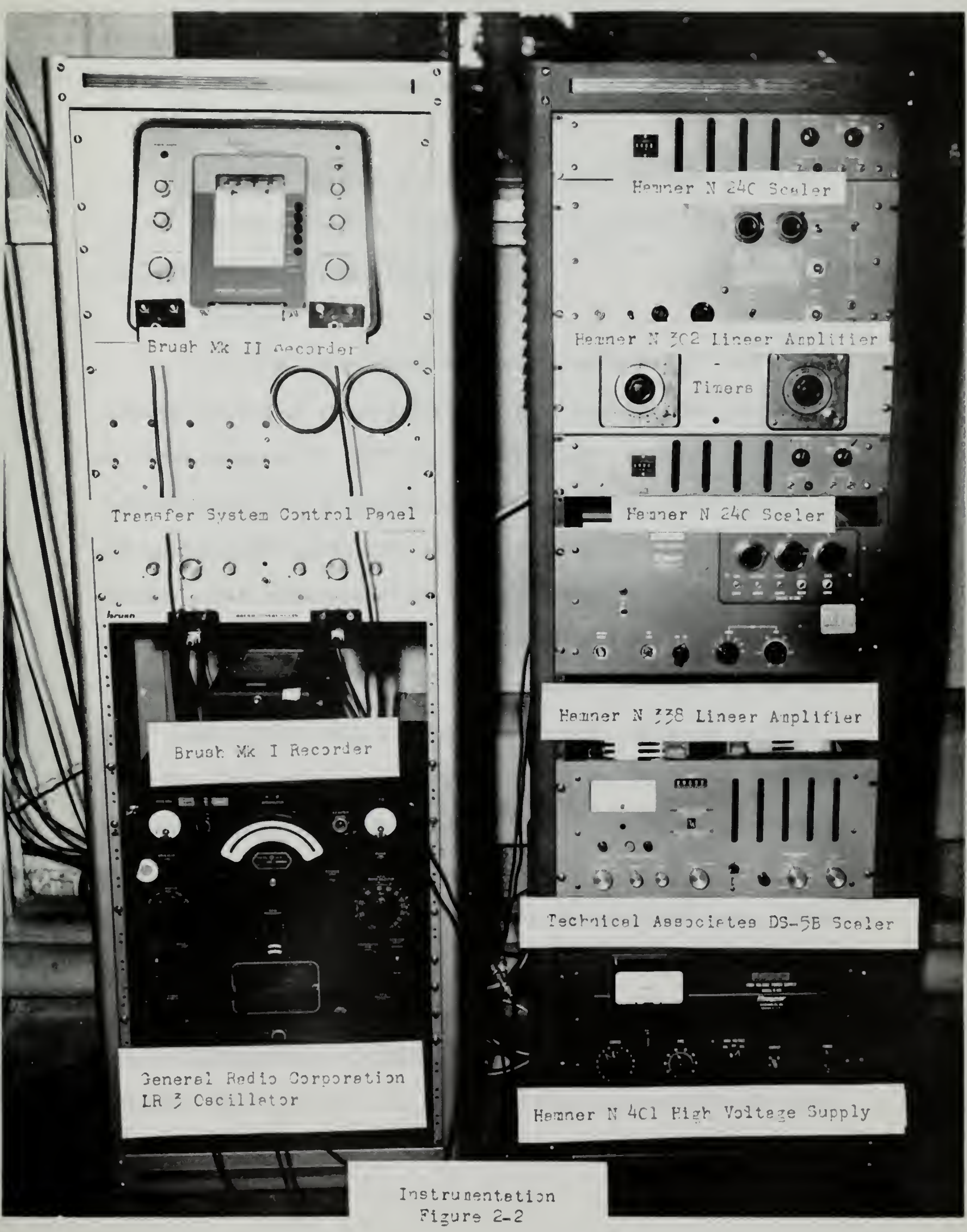


Figure 2-1. Counting System



Instrumentation
Figure 2-2

scintillation counter, in approximately one-tenth of a second. The transfer system is completely closed, allowing recycling of the sample without disturbing the detection arrangement.

b. Isotopes

Isotopes used in the investigation were highly enriched samples procured from Oak Ridge National Laboratory. Isotopic analyses are included as Appendix I.

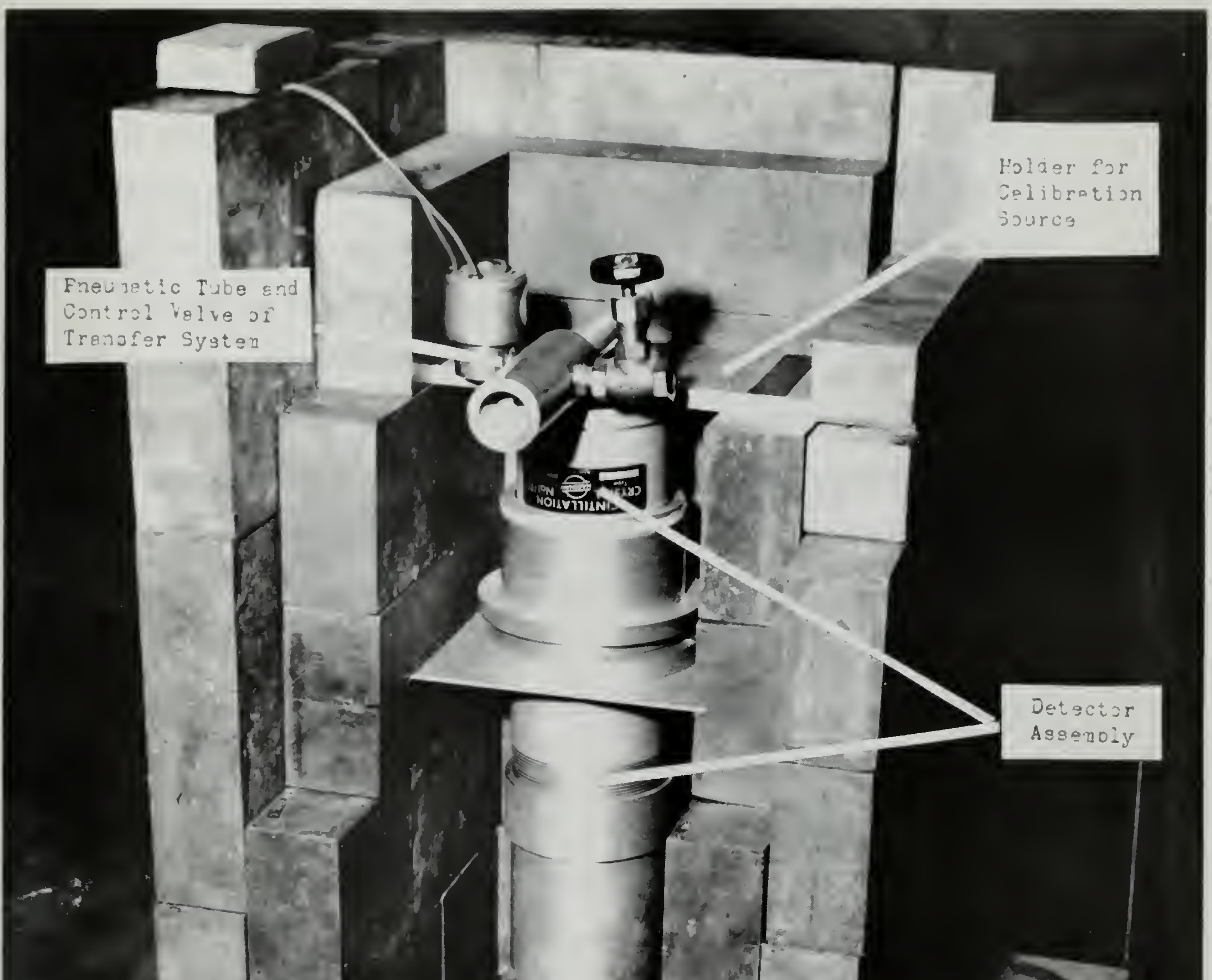
c. Detector

(1) The scintillation detector consisted of a 3 inch diameter by 3 inch thick NaI(Tl) crystal (Harshaw Type 12A12), a 3 inch diameter by 11/32 inch thick NaI(Tl) crystal (Harshaw Type 12U12-X), and a Electra Megadyne Inc. Model 9578B photomultiplier tube. The faces of the crystals are sealed by glass plates. This photomultiplier tube was rated to have a stability of less than 1% with a count rate change up to 10,000 counts per second and to be capable of a resolution of less than 9% for the Cs^{137} peak. The 3 inch x 11/32 inch crystal contained an Americium 241 alpha source mounted directly in the center of the crystal; the source material is deposited on a 1/4 inch diameter Platinum foil. Source activity was rated to be 1000 cps ± 15%, with a pulse height resolution of 10% or better for the Americium peak. These items were assembled with the thinner crystal sandwiched between the larger crystal and the phototube, and the two crystals and tube were held together and completely enclosed in a 3 inch magnesium cylinder which served as both an enclosure and mounting for the assembly. In assembling the detector, the glass interface

between the two crystals and the glass interface between the small crystal and the photomultiplier were both sealed with Dow Corning silicone grease, Compound C-20057. The peak of the 5.50 Mev alpha source yielded a counting rate of approximately 600 counts per second, resulting in 2.65 Mev gamma equivalent pulses which were used as a reference in monitoring the gain stability of the system. Resolution of the alpha peak was measured to be 13% at 975 volts. Resolution of the Cs^{137} peak with the above assembly was measured to be 10.9% at 975 volts. The entire counting assembly was shielded by a lead enclosure of 2 inches minimum thickness and was offset from the pneumatic tube center-line to reduce any increase in the background rate that might result from reactor operation. Incorporated with the detector as an integral part of its shielding, was a mechanical slide device which afforded exact repositioning of the Cesium calibration sources used in the experiment. See Figure 2-3.

(2) An "operational" pre-amplifier (Hamner 361), attached directly to the detector mounting assembly and enclosed in the shielded area, relayed pulses to the two amplifiers. (All signal pulses were transmitted over shielded cables.) Preamplifier power (260 volts) was supplied by a stabilized power supply unit in the Hamner N-338 amplifier.

(3) High voltage for the photomultiplier tube was



Counting Area, Shielding Partially Removed
Figure 2-3

provided by a Hammer N-401 High Voltage Power Supply, which provides 500-1800 volts, continuously variable, with a rated stability of 2.5 parts per million per milliampere throughout its operating range of 0-5 milliamperes at 1000 volts output. The unit was operated at 975 positive volts output, 2 ma current, and no high voltage drift was observed during any of the decay runs.

d. Analyzers

(1) A Hammer Model N-338 Linear Amplifier was used for analysis of pulses produced by the nuclides being studied. This amplifier uses double delay line pulse shaping, providing for fast baseline recovery under high duty cycle operation. Its circuitry is especially designed for use with large overload signals, counting rates up to 250,000cps, and for use on pulses having slow rise times, such as from NaI crystals; thus it was particularly suited for measuring gamma pulses below one Mev in the presence of pulses from the alpha source. The amplifier has a rated gain stability of 0.25% per day and is rated at 0.1% linearity; linearity checks performed during the experiment showed no deviation from rated linearity. The amplifier also provides for either differential or integral discriminator output, together with a separate integral discriminator output. It was operated in both integral and differential modes, with the differential mode being used to analyze Tungsten and Ytterbium

pulses, and the integral mode being used to analyze Hafnium pulses. In making special calibration runs to determine the pile-up correction factor (See Appendix IV), integral counts over the entire energy spectrum and differential counts over the energy region under investigation, were recorded simultaneously.

(2) A Hamner Model N-302 Linear Amplifier was used for analysis of the gain-monitor alpha pulses.[*] This non-overload pulse amplifier is particularly designed to amplify the output of scintillation detectors and employs input pulse clipping to reduce pulse pile-up. Linearity is rated at better than 0.2%. This unit provides for operation in either differential discrimination or integral discrimination modes. The integral mode was employed throughout the experiment. The discriminator was set at a level well above the experimental energy levels being observed, but slightly below the equivalent gamma energy level of the constant count rate alpha source. This count rate was monitored throughout all observations for a change in counting rate that would indicate a shift in gain of the detection system (See Appendix V). Periodic checks of linearity using a mercury pulser showed no deviations from rated amplifier linearity.

e. Count recording system

Outputs from the two amplifiers were fed to Hamner Model N-240 High Speed Scalers. These scalers were found to have a 1.4

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[*] Hereafter, this unit is termed the gain-monitor channel, whereas the Hamner N-338 circuit described above in (1) is termed the decay channel.

microsecond resolving time using a double pulser. They were modified so that when operated in a continuous mode, each scaler delivered one pulse for every 10, 100, 1000, or 10,000 input pulses to one of the two twin-channel continuous strip chart recorders. The recorder used to record decay inputs (Brush Mk II) was equipped with a variable chart drive with 1, 5, 25, and 125 mm per second speeds, and the recorder used for the gain monitor inputs (Brush Mk I) employed a similar drive, but with 5, 25, and 125 mm speeds only. Both recorders are capable of recording the peak-to-peak amplitude of a constant voltage sine wave of 100 cps frequency.

f. Time calibration system

To provide a precise time standard, signals from a crystal controlled oscillator with a rated frequency stability of .005% were recorded on the second channel of each of the recorders. The oscillator used, a General Radio Corporation Model LR-3, was designed as a frequency standard for radio frequency calibration, and is capable of delivering outputs of 10, 20, or 100 kc/s. The selected output was delivered to a five decade 200 kc/s Technical Associates Model DS-5B scaler. This scaler was modified so that when used with the above described outputs, it was capable of delivering pulses to both of the Brush recorders with time intervals of 1/100, 1/10, 1/20, 1/2, or 1 second. Pulses at 1/10 second intervals were used exclusively throughout this investigation.

3. Calibration

a. Dead-time and pile-up time

The overall circuit dead-time was measured by the two source method and found to be 1.9 ± 0.2 microseconds. The effective pile-up time was estimated to be 0.8 ± 0.4 microseconds. The pile-up time calculation was based on a series of measurements with Cs^{137} sources of widely varying activities, together with a simplifying approximation for the Cs^{137} pulse height spectrum.

b. Amplifier linearity

The outputs of the differential discriminators, which are incorporated into the amplifier units (See Section II), yield pulses whose amplitudes fall within the range E and $E + \Delta E$. The interval ΔE is referred to as the window width. Both E and ΔE denote dial settings whose numerical values correspond approximately to the appropriate pulse amplitudes in volts.

The linearity of the amplifiers was checked by using a mercury pulser (Hamner N-101). The amplitude of the pulser's output pulses was accurately known. Plots of pulse amplitude versus E dial setting indicated linearity within the stated rating of the amplifier units.

c. Window calibration

The window width ΔE was calibrated in terms of E dial increments by using fixed amplitude pulses from the mercury pulser. It was found that the actual window width was 0.4 units wider than the ΔE setting.

d. Energy calibration

For the amplifier unit used to analyze the decay pulses, the pulse height versus E dial-setting curve (para. b, above) passed through the origin. This simplified energy calibration. Essentially, the E dial was calibrated for energy by always adjusting the amplifier gain so that the Cs^{137} peak (0.662 Mev) came approximately at an E dial setting of 85 volts. (The E dial setting is normally referred to in terms of volts). Since the ΔE dial was calibrated in terms of the E dial, the ΔE dial was also effectively calibrated in energy. For the details of the exact E dial energy calibration see Appendix V.

e. Gain shift

The Am^{241} alpha source, which was incorporated into the scintillation counter, was used to monitor the effective gain stability of the scintillation counter. The gamma equivalent energy of the pulses from the alpha source was about 2.65 Mev. Since the isomers which were investigated all had gamma energies below 1 Mev, pulses from the alpha source peak could be effectively isolated from those of the isomer under study.

The integral discriminator of the alpha pulse amplifier unit was set at an E dial value which corresponded to the steep rising portion of the peak of the alpha source pulse height spectrum. Any change in the gain of the scintillation counter

would thus result in a relatively large change in the counting rate of the integral discriminator. Thus the latter could be used to determine the relative gain of the scintillation counter as a function of time, simultaneously with the recording of decay counts from the isomer. The details of the use and calibration of the alpha source gain shift monitor are discussed in Appendix V.

Detailed integral and differential pulse height spectral curves of the alpha source and the standard Cs^{137} source were measured. The presence of the small NaI(Tl) Crystal, which contained the constant count-rate alpha source, worsened the resolution of the scintillation counter, as expected. (See Sect. II) More than half of the alpha source activity was in the vicinity of the main peak, whereas the rest was distributed over the lower portion of the pulse height spectrum.

f. Measurements for pile-up corrections

For two of the isomers studied (Yb^{177m}, W^{183m}) the decay counts were taken from the appropriate energy (pulse height) interval defined by E and Δ E. (In the case of Hf, one set of data was taken in this way). For a particular isomer, pile-up corrections were made to the decay counts from the window ΔE, by having knowledge of the total counts arising from the pulse height spectrum in the pulse height intervals above and below the window. The above counts were obtained as a function of time by utilizing the double discriminator feature of the N-338 amplifier unit. For example,

the usual decay data was obtained from the differential discriminator. At the same time, counts were obtained from a second discriminator in the integral mode, with the same E setting used on the differential discriminator. Counts from both these discriminators were recorded simultaneously on one of the twin channel Brush recorders. A similiar type run was made to determine the counts in the spectral region below the window. All of the data from these special "calibration" runs were then effectively normalized to the window decay counts for the run whose data was being used to directly measure the half-lives of the isomeric transitions.

4. Procedure:

Prior to the actual run and with the reactor at the appropriate power, the differential discriminator of the decay channel amplifier unit was calibrated in terms of energy by using the standard Cs^{137} source. Then the appropriate energy interval for the isomer under study was transferred to the E and ΔE dial settings. The energy intervals used, together with radiation powers and exposure times* for the respective isomers, are as follows:

Yb^{177m}	20 watts	10 seconds	.09 to .172 Mev
W^{183m}	40 watts	10 seconds	.09 to .172 Mev
Hf^{178m}	6 watts	10 seconds	Integral setting

Next, each scaler was set to feed one pulse per one hundred counts into the Brush recorders, and the scaler controlling the output of the timing crystal was set to feed one pulse per one tenth second into the recorders.

After a background count was taken in the decay channel, both the decay channel and the alpha channel were connected to the Brush recorders. The recorders were started, and the capsule containing the isotope was fired into the core of the reactor. As the firing switch was thrown, the timing apparatus was set to feed one pulse per second into the recorders. After approximately ten seconds irradiation time, the capsule was fired from the reactor core into the proper counting position, and the timing apparatus was reset to

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* These neutron exposures result in initial total counting rates whose dead-time correction is approximately 8% or less.

feed one pulse per one tenth second into the recorders. (The difference in timing scales offered a means by which the irradiation time as well as the travel time for the capsule from the reactor core to the counter could be estimated.)

By this method two tapes were obtained: The first tape contained counts from the decay of the radioactive isomer along with an accurate time record; the second tape contained counts from the constant count alpha source, along with the same time base, and was used to check gain shift. See Sect. 3 and Appendix V.

If more than one run was to be made, in all cases except Yb^{177m}, the next run was not started until the activity from the previous run had decayed to such a point that it was indistinguishable from background. Also, the gain was checked to ascertain whether or not the discriminator settings had to be changed.

In the case of Yb^{177m}, a daughter decay was present (Yb^{177m} to Lu^{177m}, half-life 1.9 hrs). The Lu^{177m} is also radioactive and has a half-life of 6.8 days. Therefore, the above procedure had to be changed. It was predicted by use of chain decay rules that within approximately five minutes after the first run, the background activity would be 4.8 counts per second and would remain essentially constant over the period of time necessary to complete another run.

Experimental observations showed that the activity (depending on the reactor power and the irradiation time) was 5.1 counts per second above background and decreased 6.5 % of this value in ten minutes.

Therefore, since a Yb^{177m} run lasted approximately two minutes, it was possible to wait five minutes between runs and then take an activity reading which, because of the short time interval of the run and the apparently slow decay, could be used as an effective background in the treatment of data.

could [illegible].

5. Data Reduction and Analysis

The data from the experimental observations were recorded on the tapes of two different Brush recorders, with one tape containing data from the constant count-rate alpha source and the other tape containing data from the isotope under investigation. Each tape was a twin-channel strip chart on which one channel contained blips indicating the number of gamma pulses detected, in increments of 10, 100, 1000, or 10,000 pulses; and the other channel contained time pulses, with each pulse representing one-tenth of a second. See Figure 5-1.

a. Decay data

Before extracting decay data, the time channel of each tape was analyzed. By correlating increments of tape length with the timing pulses, it was possible to determine the rate of tape flow to within one-one hundredth of a millimeter per second. Some tapes were found to have speed variations greater than 1/10mm per second. These were either discarded or the appropriate time interval was extracted directly from the timing pulser instead of using the reduction technique described below. Maximum error in timing interval using this technique is estimated to be 0.5%, as compared to less than 0.1% for those tapes determined to have a constant flow rate.

The decay channel data of the tapes were reduced as follows: A time increment, of approximately 3/10 of the half-life under

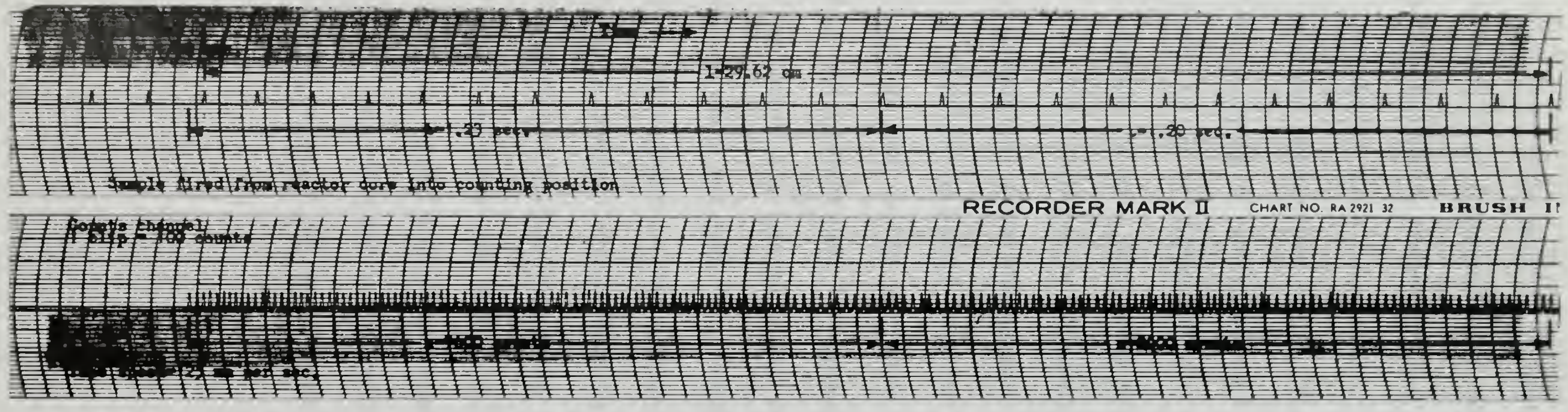

Figure 5-1

Sample Data Tape

study, was determined. (The error in true counting rate resulting from use of this time increment is 0.18%) The entire decay tape was then subdivided into approximately equal intervals of length (based on the previously determined tape flow rate) corresponding to this time increment, with each time increment taken to include complete decay-count increments only. This procedure avoided interpolation of the randomly distributed count increments and allowed all interpolation to be performed on the linear time increments. The number of counts in each increment and the length of each increment on the decay tape were determined.

This information, plus background and tape speed were used as inputs for a digital computer program. The output of this program consisted of raw counts per time interval, counting rate (both corrected and uncorrected for background), beginning time of each counting interval, and mid-time of each time interval. A sample output of this program is included as Appendix II. Portions of this data could be used directly for plotting; the other portions were used as input data for the Frantic Program for Analysis of Exponential Growth and Decay Curves [8]. This program has been written to process raw counting data and fit to these data, by the least-squares techniques, equations for multiple exponential growth and decay. The program requires inputs of raw counts, counting interval, duration of the counting interval and uncertainty

thereof, background, dead-time and uncertainty thereof, and time and time uncertainty. Among the outputs of the program are half-lives and standard deviations thereof, decay constants and standard deviations thereof, initial activities, the Chi-square value and variance-of-fit for use in goodness-of-fit tests, and a histogram of the deviations of the final calculated points from the data points.

Using the Frantic Program, the Tungsten and Ytterbium isomers were analyzed as single component decays, with a measured background as an input. The Hafnium sample was analyzed as a three component decay: Hf^{178m},Hf^{179m},and a computer determined background level. A sample output of the Frantic Program is included as Appendix III.

The method of determining time of commencement of counting intervals, and durations of counting intervals have been discussed above. Background was measured directly over the energy interval under investigation immediately before each run and was assumed constant during the counting interval. Typical background rates encountered in the energy regions in which measurements were taken were 1300 counts per second while taking Hafnium measurements (integral counts over the entire energy spectrum, including all pulses from the alpha source), and thirty counts per second while taking Ytterbium and Tungsten measurements (differential counts

over 0.085 Mev range). Irradiation of a dummy projectile (constructed in exactly the same way and of the same materials as the projectiles used in the experiment, but containing no sample) under all radiation powers and times used in the investigation indicated no significant increase over background rates.*

Time uncertainties were calculated from the estimated accuracy with which the length of tape representing a counting interval could be measured, rate of tape flow, and the average length of the time intervals used for a particular measurement.

The method of determining raw counts discussed above was adequate for those runs in which the counts over entire energy spectrum were recorded. In these runs dead-time corrections could be made by the Frantic Program, if the correct dead-time was inserted as a program input. For those cases in which only a portion of the energy spectrum was investigated, counting data had to be corrected to compensate for counts lost due to the dead-time of the decay channel circuit and for the phenomenom of "pile-up", which may cause either a loss or gain in the total counting rate, depending upon the counting mode used to measure the decays. (These problems are discussed more thoroughly in App. IV).

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* Maximum effect was observed when the dummy was radiated at the same power used for Tungsten radiation. Background at operating power was measured to be 35 counts per second; when the "hot" dummy was fired into counting position, the counting rate increased to 45 counts per second and dropped back to 35 counts per second within 1.2 seconds. Initial activities for Tungsten during experimental observations were approximately 8,000 counts per second.

Raw count data was corrected for the above effect by adding or subtracting counts to give a revised raw count which was used as an input into the Frantic Program.

b. Gain-shift data

The tapes of the alpha counts were analyzed in a similar but simpler manner. Each tape was subdivided into periods of approximately one second, rounding off each period so that only complete count increments were included. Rounding off in this manner allowed all interpolation to be performed on the periodically occurring time blips rather than on the randomly occurring gamma pulses. Count rate for each interval was determined by dividing the counts in an interval by the duration of the interval. This count rate was then plotted against time to determine whether any change in counting rate, indicating a shift in gain, had occurred.

6. Results

a. Results of the half-life determinations of individual observations and the average values for each isotope are as follows:

(1) Ytterbium (Yb^{177m})

6.384 ± .039 sec.

6.417 ± .038

6.386 ± .037

6.410 ± .024

Average value: 6.40 ± .02 sec.

(2) Tungsten (W^{183m})

5.120 ± .039 sec.

5.156 ± .025

5.153 ± .027

Average value: 5.14 ± .02 sec.

(3) Hafnium (Hf^{178m})	(Hf^{179m})
3.87 ± .11 sec.	18.39 ± .33 sec.
3.88 ± .13	18.27 ± .63
3.63 ± .11	17.89 ± .46
Average value: 3.79 ± .07 sec.	Average value: 18.18 ± .28 sec.

b. These results are in agreement with the results published in references [4], [6], and [9]. These publications list values of 6.5, 6.4, and 6.0 seconds respectively for Yb^{177m}; 5.3, 5.5, and 5.5 seconds respectively for W^{183m}; 4.3, 3.5, and 4.8 seconds respectively for Hf^{178m}; and 19.0, 18.6, and 19.0 seconds respectively for Hf^{179m}.

The uncertainties listed with each of the above half-life measurements are standard deviations resulting from computer calculations. These calculations are based on uncertainties in count rate, background, dead-time, and counting interval. The agreement between individual measurements is seen to be essentially within statistical uncertainty. The uncertainties listed with the average half-life values are the standard deviations of the mean values.

The overall uncertainty of the average half-life values for Yb^{177m} and W^{183m} taking into account systematic errors (see para. c., d., e., following) should be about 1%. The half lives of Hf^{178m} and Hf^{179m} were determined by irradiating a sample containing 84.52% of Hf^{177m} and 9.2% of Hf^{178m}. Additional measurements are necessary before an overall uncertainty can be estimated for their half-lives.

c. On most of the gain monitor tapes which were examined, the variations in counting rates were indistinguishable from statistical fluctuations. On one of the Tungsten runs, however, an overall decrease in gain of approximately 0.25% was observed. The assumption was made that the gain decreased linearly with time over the period of decay measurement. When corrections to the data were made on this basis, the computer program indicated a change in half-life value from 5.12 seconds to 5.09 seconds, a change of 0.6%. Since gain shifts of the order of 0.25% were readily discernible from analysis of gain-monitor data, it is believed that 0.6% represents a good estimate of the maximum error due to gain shift.

d. Corrections for dead-time and pile-up effects were made as outlined in Appendix IV. The magnitude of error resulting from these effects was estimated by analyzing data from runs in three ways, depending on the assumptions made for dead-time and pile-up corrections. The runs were first analyzed using the best estimates of all corrections factors to determine the values reported above. These same data were then analyzed without making either dead-time or pile-up corrections; the maximum difference between these values and the reported values was 0.8%. The analyses were then repeated twice using values of pile-up time that were 50% above and below the best estimated value; the maximum difference between these half-life values and the reported values was 0.7%. It was also noted that the computer-determined variance-of-fit and Chi-square values were significantly better for those analyses of Tungsten and Ytterbium data in which best estimate corrections were made. This supports the assumptions that were made in evaluating the dead-time and pile-up corrections.

e. The Hafnium runs whose results are reported in paragraph a. were analyzed by using the measured background counting rate as an estimate for the computer program input. The computer program then determined that value of constant background counting rate which gave the best least squares fit to the data. The difference between this background value and the measured value (about 3 %) may well be due to the presence of long-lived contaminating activities.

Further checks on the reported results were obtained by analyzing Hafnium data taken with the discriminator set well above the highest known gamma energy resulting from Hf^{179m} decay; the analysis of these data yielded a half-life for Hf^{178m} of 3.86 ± .04 seconds. A similar check on the Yb^{177m} and W^{183m} results was obtained by analyzing data taken over the entire energy spectrum (integral counts). The results of these data, which were taken under less favorable conditions than the normal runs relative to timing accuracy, were within 1% (6.47 ± .14 sec.) and 4% (5.36 ± .17 sec.) respectively of the reported values for the half-lives.

f. The results obtained in this investigation should represent significant improvement in precision over presently published values of the half-lives of the isomers investigated. The equipment and techniques presently employed should yield good results when applied to other isomers with even shorter half-lives. However, for measurements of half-lives less than 5 seconds, it would be desirable to incorporate automatic controls for the pneumatic sample transfer system presently in use. Further improvements in accuracy should result from the use of a faster scintillation detector and analyzer system, and from the use of a reliable multi-channel analyzer for better energy calibration and pile-up correction.

APPENDIX I

ISOTOPIC ANALYSES

(As reported by Oak Ridge National Laboratories)

1. Tungsten:

Isotope	Atomic Percent	Precision
180	less than .05	---
182	94.4	±0.1
183	2.53	0.05
184	2.32	0.05
186	.8	0.05

2. Hafnium:

Isotope	Atomic Percent	Precision
174	less than .05	---
176	1.15	± .05
177	84.52	0.1
178	9.2	0.05
179	2.22	0.05
180	2.91	0.05

3. Ytterbium:

Isotope	Atomic Percent	Precision
168	less than 0.01	---
170	.03	---
171	.16	0.02
172	.29	0.02
173	.29	0.02
174	1.45	0.05
176	97.77	0.1

[illegible]

[illegible]

[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]
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[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]

```
        PROGRAM  DATA
C       REVISE BACKGROUND AND SPEED CARDS FOR EACH DATA RUN
C       TUNGSTEN NO 2   22 MAR 65   11.71 SEC AT 40 WATTS
        TIME = 0.0
        SPEED =12.5004
        BGRND =30.32
     12 READ 10,COUNTS,CM,Q
     10 FORMAT(2F15.3,I2)
        PERIOD = CM/SPEED
        RATEMP = COUNTS/PERIOD
        RATE = RATEMP- BGRND
        PDHALF=PERIOD/2.0
        TIMIDL= TIME+ PDHALF
        PRINT 11,TIME, PERIOD, COUNTS, RATE, PERIOD, RATEMP, TIMIDL
     11 FORMAT(1P3E15.5,0P4F12.4)
        TIME=TIME+PERIOD
        IF  (Q) 12,12,14
     14 STOP
        END
        END
     .00000E-01    1.59915E+00    1.61000E+04   10037.5359        1.5991  10067.8559         .7996
    1.59915E+00    1.60395E+00    1.31000E+04    8137.0236        1.6039   8167.3436        2.4011
    3.20310E+00    1.59115E+00    1.03000E+04    6442.9892        1.5911   6473.3092        3.9987
    4.79425E+00    1.59275E+00    8.30000E+03    5180.7960        1.5927   5211.1160        5.5906
    6.38700E+00    1.59275E+00    6.80000E+03    4239.0281        1.5927   4269.3481        7.1834
    7.97974E+00    1.60235E+00    5.40000E+03    3339.7329        1.6023   3370.0529        8.7809
    9.58209E+00    1.58635E+00    4.30000E+03    2680.3063        1.5863   2710.6263       10.3753
    1.11684E+01    1.58395E+00    3.50000E+03    2179.3467        1.5839   2209.6667       11.9604
    1.27524E+01    1.56875E+00    2.80000E+03    1754.5408        1.5687   1784.8608       13.5368
    1.43211E+01    1.59355E+00    2.40000E+03    1475.7523        1.5935   1506.0723       15.1179
    1.59147E+01    1.54635E+00    1.90000E+03    1198.3794        1.5464   1228.6994       16.6879
    1.74610E+01    1.53915E+00    1.50000E+03     944.2434        1.5392    974.5634       18.2306
    1.90002E+01    1.53595E+00    1.20000E+03     750.9550        1.5360    781.2750       19.7682
    2.05361E+01    1.45995E+00    9.00000E+02     586.1381        1.4600    616.4581       21.2661
    2.19961E+01    1.48875E+00    8.00000E+02     507.0427        1.4888    537.3627       22.7405
    2.34848E+01    1.62235E+00    7.00000E+02     401.1534        1.6223    431.4734       24.2960
    2.51072E+01    1.63755E+00    6.00000E+02     336.0816        1.6375    366.4016       25.9260
    2.67447E+01    1.53995E+00    5.00000E+02     294.3657        1.5400    324.6857       27.5147
   STOP
 TIME,  0 MINUTES AND  27 SECONDS
```

Appendix II

Data Reduction Program

```
                    YTTERBIUM NO 3 26 FEB 65  1C.6 SEC AT 10 WATTS,INCL PILE-UP AND DEAD TIME CORR

     YB 3 26FEB = FRANTIC IDENTIFICATICN                             .00000 = TAU (TIME TC COUNTING)
              1 = NUMBER OF COMPCNENTS                               .00000 = TAUD (DEAD TIME FACTOR IN MICRO TIME UNITS)
              0 = NUMBER OF PARAMETERS HELD FIXED                    .00000 = ERRCR IN DEAD TIME FACTOR (MICRO TIME UNITS)
             16 = NUMBER CF CATA POINTS                            40.58CCC = BACKGROUND
             14 = DEGREES OF FREEDOM                                 .01000 = UNCERTAINTY IN TIMING INTERVAL IN TIME UNITS
              7 = ITERATIONS                                        1.00000 = DATA SCALE FACTCR
              2 = STATISTICAL WEIGHTS
+                                                                   1.00CCC = NCRMALIZATICN FACTOR
              0 = NUMBER OF POINTS (*) DEVIATING MORE THAN 2 SIGMA
     2448.08062 = LEAST SQUARES CETERMINANT
         .0C049 = DELTA VARIANCE OF FIT X 1C(6)
         .50543 = WEIGHTED VARIANCE OF FIT
        8.52702 = CHI SQUARE


      A(ZERO)        SIGMA        LAMBDA         SIGMA       N(CRIGINAL)      SIGMA      HALF LIFE      SIGMA

      9123.587       40.444     .108127350     .COC398450      84378.          486.        6.4105        .0236
         ORIGINAL ESTIMATES

      9105.722                  .1079000C0

                                  ANALYSIS OF THE DEVIATIONS

         0     0    0    0    0    0    C    0    0    5    3    4    3    1    0    0    0    0    C    0    0    C
SIGMA -XS  -4.5 -4.0 3.5 -3.0 -2.5 -2.0 -1 .5 -1.0 -0.5 -0.0  C.C  0.5 . 1.0  1.5  2.0  2.5  3.0  3.5  4.0  4.5  XS


        BEGINNING    INTERVAL      ORIGINAL      CORRECTED      CALCULATED     INSTANTANEOUS    WEIGHTS        DELTA
          TIME         TIME         COUNTS          DATA           RATE            RATE         X 1C(3)         RATE

    1       .000       1.928       15936.1       8225.032       8235.371       8220.477         .16270       -10.339
    2      1.928       1.915       13013.7       6755.086       669C.224       6678.287         .20707        64.861
    3      3.843       1.920       10542.1       545C.097       5437.517       5427.765         .27036        12.580
    4      5.763       1.919        8491.4       4384.329       4418.367       441C.451         .34981       -34.038
    5      7.682       1.893        6859.3       3582.928       3595.314       3589.045         .43440       -12.386
    6      9.574       1.908        5638.4       2914.556       2927.858       2922.672         .55249       -13.3C2
    7     11.480       1.934        4625.5       2351.095       2379.341       2375.C11         .7C893       -28.245
    8     13.410       1.905        3715.5       1909.814       1934.118       1930.703         .86960       -24.304
    9     15.320       1.920        3109.9       1579.160       1571.994       1569.175        1.06846         7.165
   10     17.240       1.917        2606.2       1318.940       1277.490       1275.206        1.28097        41.450
   11     19.160       1.944        2103.9       1041.673       1C36.535       1034.629        1.64316         5.138
   12     21.100       1.968        17C2.4        824.461        839.343        837.762        2.08553       -14.883
   13     23.070       1.910        1401.3        693.085        680.369        679.161        2.38028        12.716
   14     24.980       2.018        1200.7        554.415        55C.309        549.219        3.08979         4.1C6
   15     27.000       1.797         900.4        460.477        447.466        446.763        3.23429        13.012
   16     28.800       2.044         80C.2        35C.907        363.609        362.870        4.65004       -12.702
TIME,  1 MINUTES AND  13 SECCNCS
```

Appendix III

Sample Computer Output

APPENDIX IV

Dead-time and Pile-up corrections

1. Dead-time

A given pulse-detecting circuit has a characteristic time (dead-time) following a registered pulse during which the circuit will not respond to another pulse. Two pulses entering the circuit within this time interval are registered as a single pulse. Under these circumstances, some counts will fail to register, and the recorded number of pulses will be lower than the true number of detector pulses. In the circuitry used in this investigation, the dead-time of the detecting circuit has been found, using the two-source method, to be 1.9 ± 0.2 microseconds.

2. Pile-up

If two gamma rays are detected by the scintillation counter within a sufficiently small time interval, the amplifying circuit is unable to distinguish between them and treat them as separate pulses. Instead, a single pulse is formed by the amplifier, whose amplitude is the sum of the amplitudes of the two individual pulses. The process by which this occurs is denoted as "pile-up", and the time interval in which it occurs is called the "pile-up time". The pile-up time, based on experimental observations using several Cs^{137} sources whose activities ranged from 2,000 counts/second to 33,000 counts/second, was determined to be 0.8 ± 0.4 microseconds. The large uncertainty is assigned to the pile-up time because the exact energy spectrum of Cesium was not used in the pile-up time calculation.

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3. Corrections for a single channel analyzer

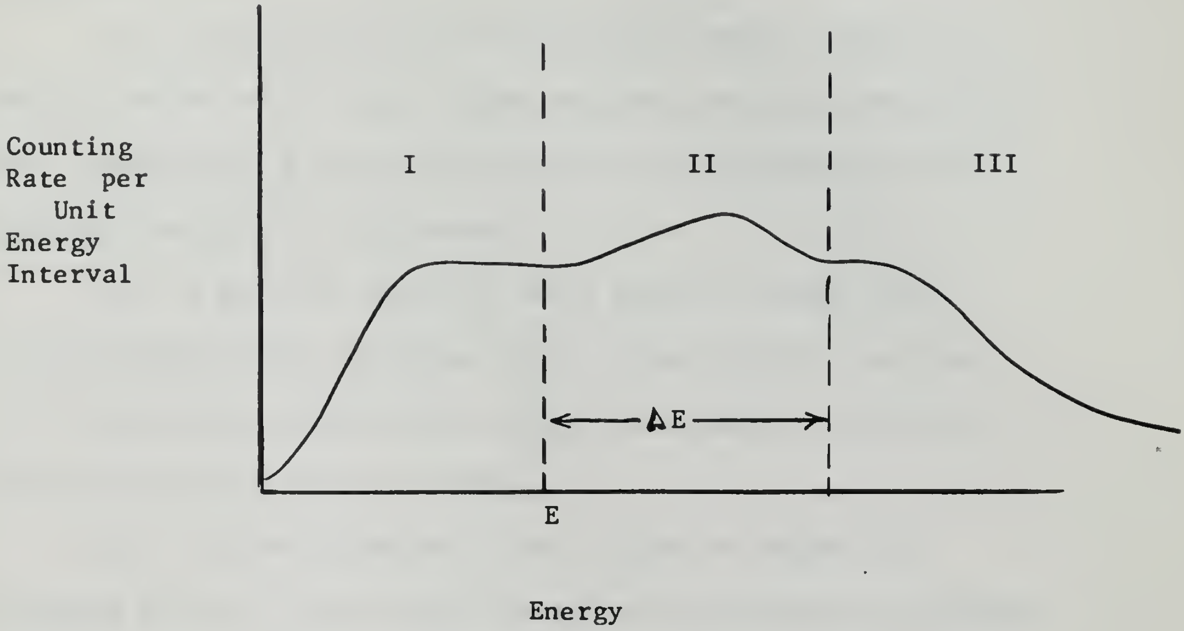

Figure IV-1

Assume an observed energy spectrum, as shown schematically in Fig. IV-1, of pulses coming from the scintillation counter at a particular instant of time. Pulses whose energies fall within the energy interval ΔE (Region II) will be recorded as the observed counting rate. It is desired to correct the observed counting rate for all possible effects due to both dead-time losses and pile-up effects.

a. Pulses fail to register in energy interval ΔE for the following reasons:

(1) Two pulses which would normally fall in region II but which have a time interval between then less than the dead-time

and greater than the pile-up time, register as one pulse. Thus, one pulse is lost.

(2) Two pulses each of which would normally register in region II which occur within a time interval less than the pile-up time, combine into a single pulse which falls outside region II [*]. Thus, neither pulse is registered.

(3) A pulse in region II and a pulse in region I (of sufficient amplitude) occurring within a time interval less than the pile-up time, combine into a single pulse which falls outside region II. Thus,one pulse is lost.

(4) A pulse in region II and a pulse in region III occurring within a time interval less than the pile-up time combine into a single pulse which falls outside region II. Again, one pulse is lost.

b. Extra pulses are registered in region II by virtue of pile-up when two pulses in region I, the sum of whose amplitudes is greater than E and less than E + ΔE, occur within a time interval less than the pile-up time. Thus, a pulse is gained.

c. For a given spectrum the proper calculation of the above five effects could be used to make an exact correction to the observed counting rate. In this experiment, the spectra involved were not accurately known, particularly since their shapes change

---

[*] Provided ΔE is less than E

rapidly as a function of time when more than one radioactive nuclide is present. However, the corrections (either exact or approximate) depend directly on the total counting rates within each of the energy regions, I, II, and III. Each of these counting rates was determined experimentally as a function of time for each of the isotopes studied, and the corrections were calculated on the basis of the simplifying assumption that the energy spectrum in each of the regions is constant throughout that region.

APPENDIX V

GAIN SHIFT

For this investigation, knowledge of the overall gain instability of the scintillation counter detection system during the half-life measurements was considered essential. Instability results from gain shifts in the scintillation counter and the amplifier-analyzer unit that follows it. Factors such as temperature variation, line voltage fluctations, and aging produce gain shifts in the scintillation counter as well as in the electronic circuitry. Relative to the time necessary to make a half-life determination, the above mentioned factors produce instabilities which are essentially long term.

However, scintillation counters are also subject to short term gain shifts caused by changes in counting rate. The amplifier and analyzer are presumably stable relative to counting rate changes. Since the counting rates change greatly in our measurements, the short term gain shift is of particular concern.

Both the scintillation counter and the associated electronic equipment used in these measurements were chosen because of their high stability in all respects.

In the discussion that follows, methods of monitoring both the long and short term gain shifts of the gamma detection system are described. Also, the method used for the energy calibration of the decay channel analyzer is described.

[illegible]

[illegible]

[illegible]

[illegible]

[illegible]

The constant energy peak from a long-lived emitter (Both alpha and gamma emitters were used.) was used to monitor gain shift. Given a known energy peak occuring at a given discriminator voltage in the differential pulse height spectrum for a given gain setting G_o, one can adjust the E dial setting to a value E_1 which occurs on the lower steep rising portion of the peak. Operation in the integral mode yields only those counts which occur above E_1. (See Figure V-1)

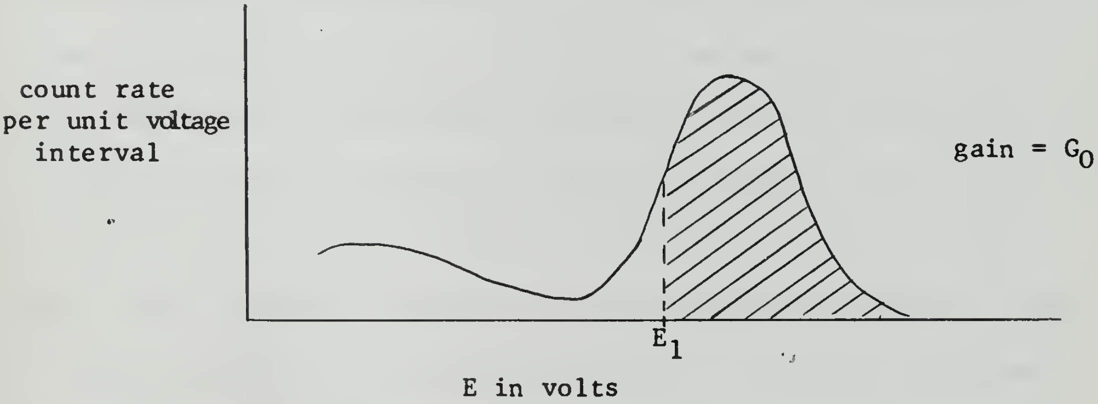

Figure V-1

As the gain varies, the energy peak shifts from its original position relative to E_1, and the observed counting rate ($\dot{n}$) varies accordingly. The variation of $\dot{n}$ with gain is the greatest when E_1 is on the steep portion of the peak. The integral counting rate at a given E_1 can be determined as a function of gain by means of a gain setting dial (whose numerical value is proportional to the amplifier gain) on the amplifier unit. From these data a calibration curve of gain-setting versus counting rate can be constructed. Relative gain can thus be determined for the system by measuring the integral counting rate at E_1 and referring to the above calibration curve.

Consider the integral pulse height spectrum for the above source at the gain setting G_o. This spectrum can be constructed by integration of the curve in Figure V-1, or by direct measurement using the integral discriminator. It can be shown that the counting rate measured at E_1 for some gain setting G, should be equal to the counting rate on the above mentioned integral pulse height spectrum (for gain equal to G_o) at a dial setting of E_1/g, where $g = G/G_o$. The solid curve in Figure V-2 represents the relative counting rate (normalized to the counting rate at G_o) versus gain-setting curve as measured experimentally. The dotted curve was predicted from the integral pulse height spectrum measured at a gain setting G_o. Incomplete agreement between the curves is probably due to instabilities that were present while the calibration data was being taken. Best agreement occurs for g in the range of 0.987 to 1.040.

Another related method of monitoring gain is based on the use of the differential discriminator. Here, the window is positioned on the lower steep rising portion of the energy peak as shown in Figure V-3.

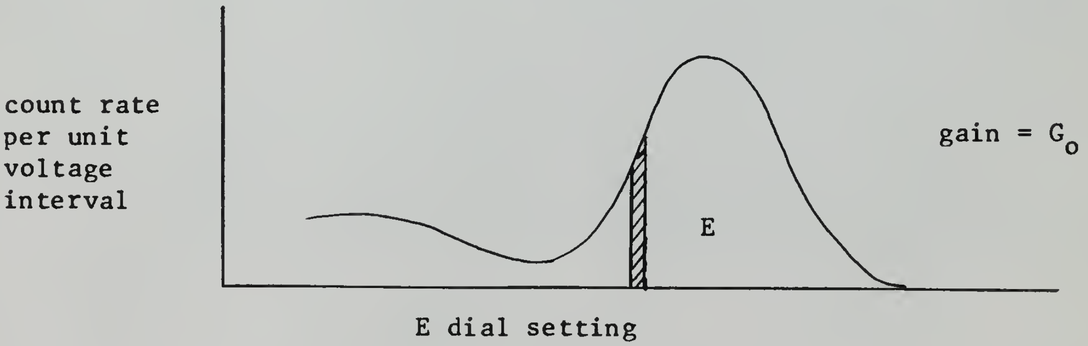

Figure V-3

$\frac{\text{counts at gain} = bG}{\text{counts at gain} = G_o}$ versus gain setting

for the alpha source

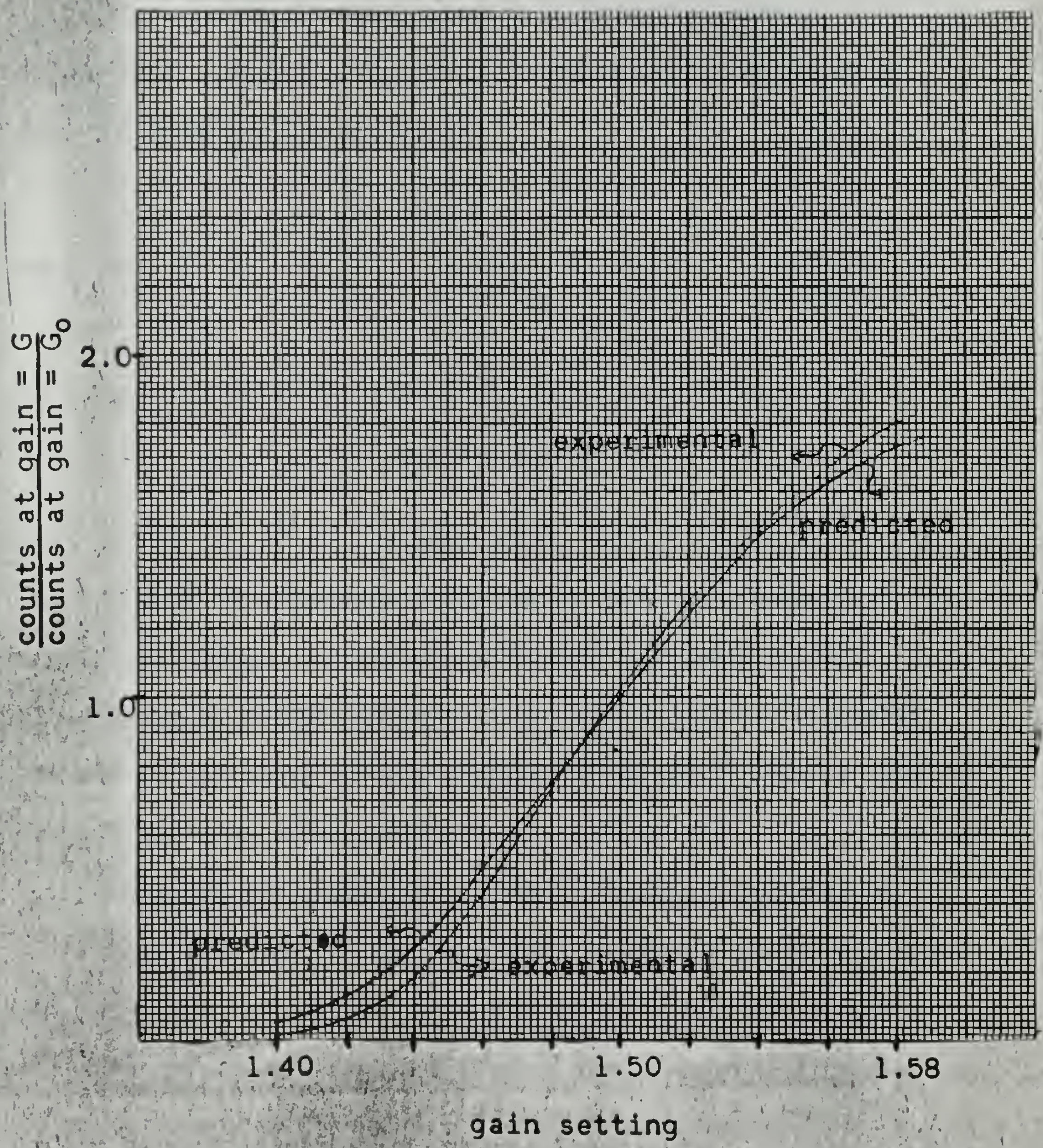

fig. V-2

Since the interval is on the steep portion of the peak, a small change in gain represents a large change in counting rate. Again, a change in gain to some value G will shift the peak relative to the window. Figure V-4 shows a typical calibration curve of counting rate versus gain setting which is normally obtained directly by measurements. It could also be obtained by proper analysis of the spectrum shown in Figure V-3.

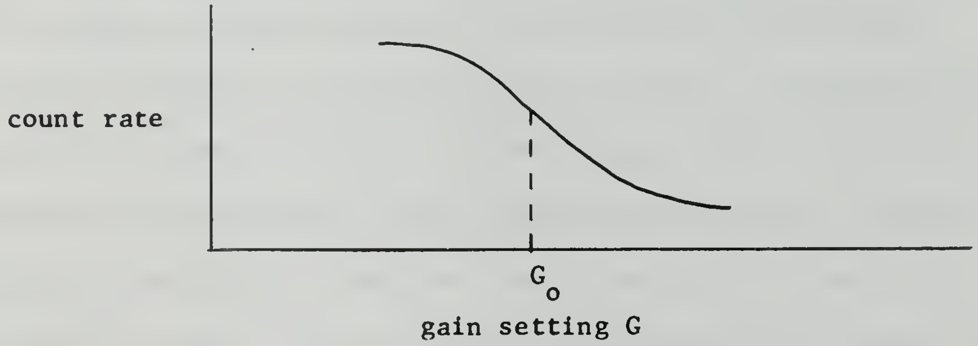

Figure V-4

The aforementioned methods were used in this experiment to accomplish two things: to monitor the short term gain shifts during an actual half-life determination; and to provide an energy calibration of the decay channel analyzer which could also be used to correct for any long term gain shifts.

a. Monitoring gain shift during the half-life determination

In order to monitor the gain shift during an actual half-life determination, it is necessary to use a constant activity source whose energy peak pulses are much greater than the gamma energy pulses associated with the isomeric decays being measured. It is

also desirable to have most of the counts associated with the constant source in the vicinity of the energy peak. Fulfillment of these requirements prevents overlap of the energy peaks and permits almost complete separation of the counts from the two sources of activity.

To accomplish this, an Am^{241} constant activity alpha source with a gamma equivalent energy of approximately 2.65 Mev., was installed internally in the scintillation counter. The gain of the amplifier unit was adjusted so that the energy peak occured at a standard E dial setting E_o. Then a gain versus integral counting rate at E_1 (such that E_1 was less than E_o and on the steep rising portion of the peak) curve was measured and plotted (see Figure V-5). The integral mode of operation was used in order to give better statistics by giving a large counting rate. Short range gain shifts occuring during the half-life determination could thus be observed by recording the integral counting rate of the standard alpha source at the E dial setting E_1, simultaneously with the counts from the decay under study.

The fact that the energy of the Am^{241} peak is so much greater than the gamma energies of the transitions under study makes it impractical to use one amplifier and scaler unit to make both observations. Therefore, it was necessary to use two such units (See Section 2). This introduced the problem of one unit experiencing a gain shift relative to the other unit. The effect of the relative instabilities of the two amplifier units should be small,

alpha source counts at E_1 = 79 volts
versus gain setting

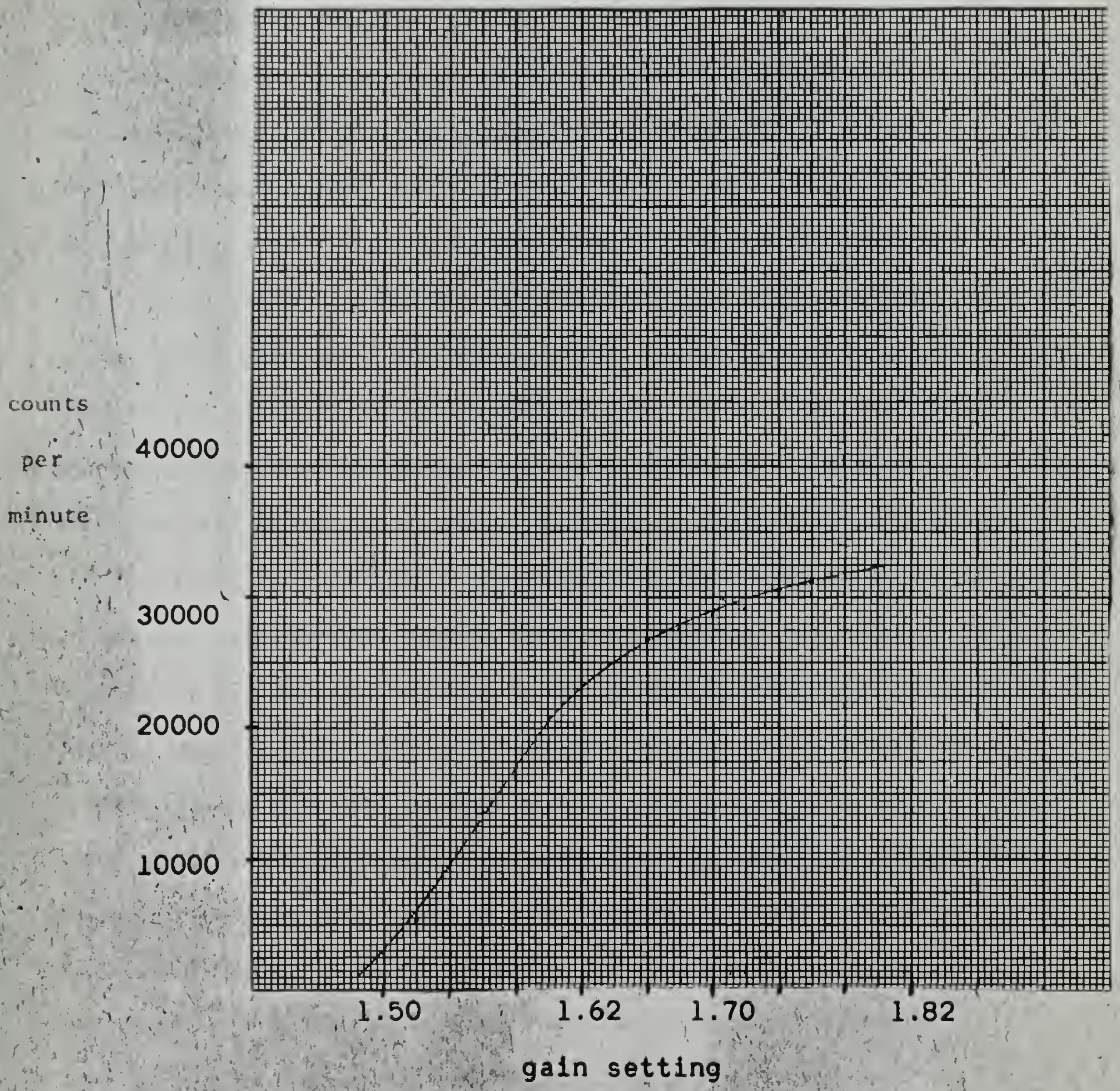

fig. V-5

because of the high sensitivity of this method for measuring gain shift. This was borne out by agreement of gain shifts as measured simultaneously by gain monitors on each of the two amplifier units.

b. Energy calibration and long-range gain shift corrections

The gamma energy peak associated with a standard Cs^{137} source was used for the energy calibration of the decay channel amplifier unit (N-338). Initially the differential pulse height spectrum for a gain setting G_o was measured. The position of the peak occured at E_o. A window ΔE was positioned at E_1, on the steep rising portion of the energy peak. The differential counting rate versus gain setting was then measured. The results are shown in Figure V-6. The approximately linear portion of this curve could be used in the usual manner as a direct measure of gain shift.

Since E_o corresponds to the known gamma energy (0.662 Mev.) associated with the Cs^{137} energy spectrum, the entire E dial (including E_1) was calibrated in terms of equivalent gamma energy. Thus, $E = E_o b/0.662$, where E is the dial setting corresponding to a gamma energy b in Mev. (assume a gain setting G_o). For some other gain G, the corresponding calibration equation is $E = gE_o b/ 0.662$ where $g = G/G_o$ is determined by a measurement with the gain shift monitor set at E_1 and ΔE. Thus the equation $E = fb$ could be used, where $f = gE_o/ 0.662$ is a known function of counting rate. A plot of f versus counting rate is shown in Figure V-7.

In practice then, the gain setting dial of the amplifier was adjusted to get approximately the standard counting rate at E_1.

Cs^{137} counts at E = 82 volts, E = 1 volt versus gain setting (G_o = 0.90)

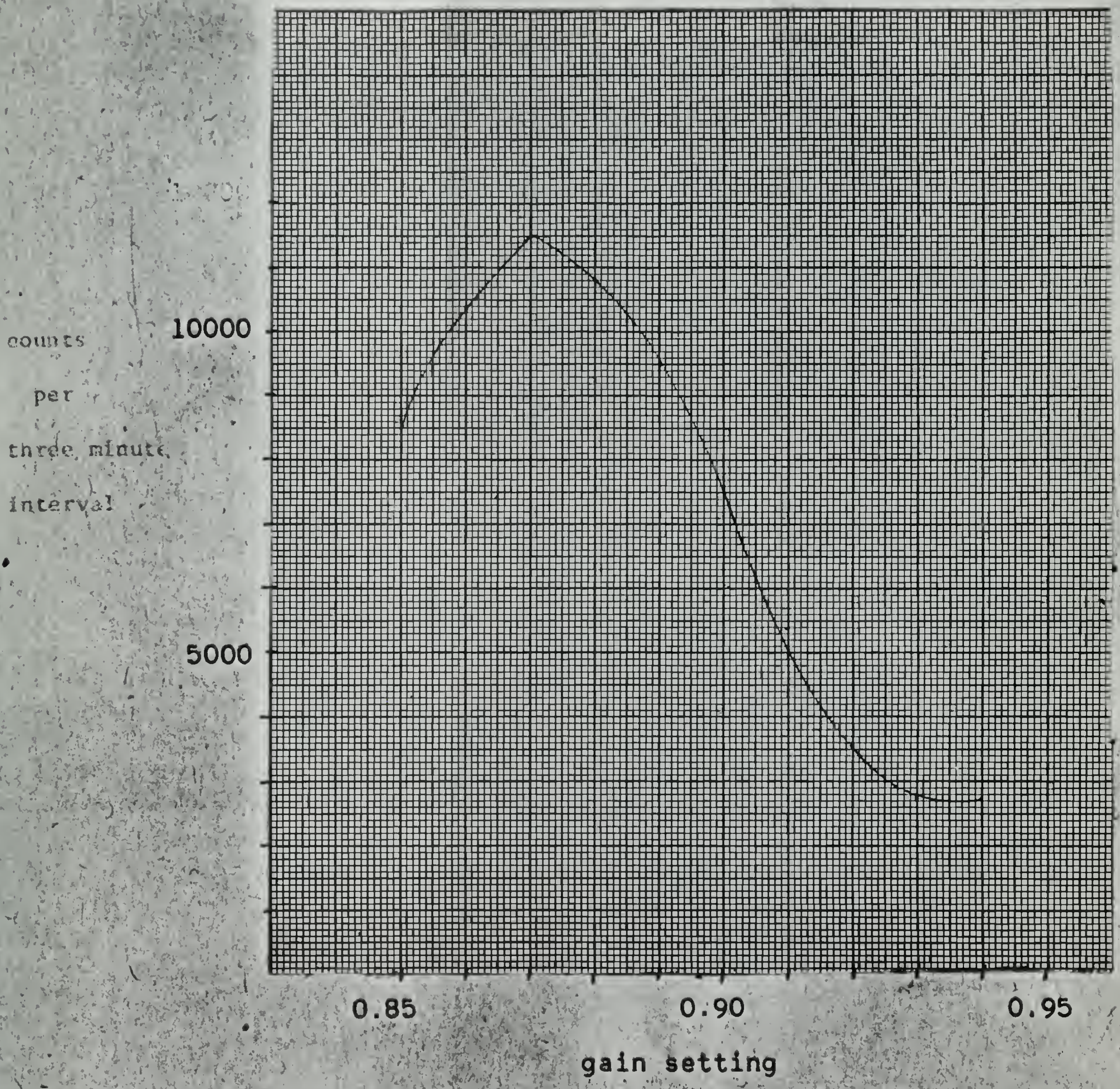

fig. V-6

f versus Cs^{137} counts at E = 82 volts

ΔE = 1 volts

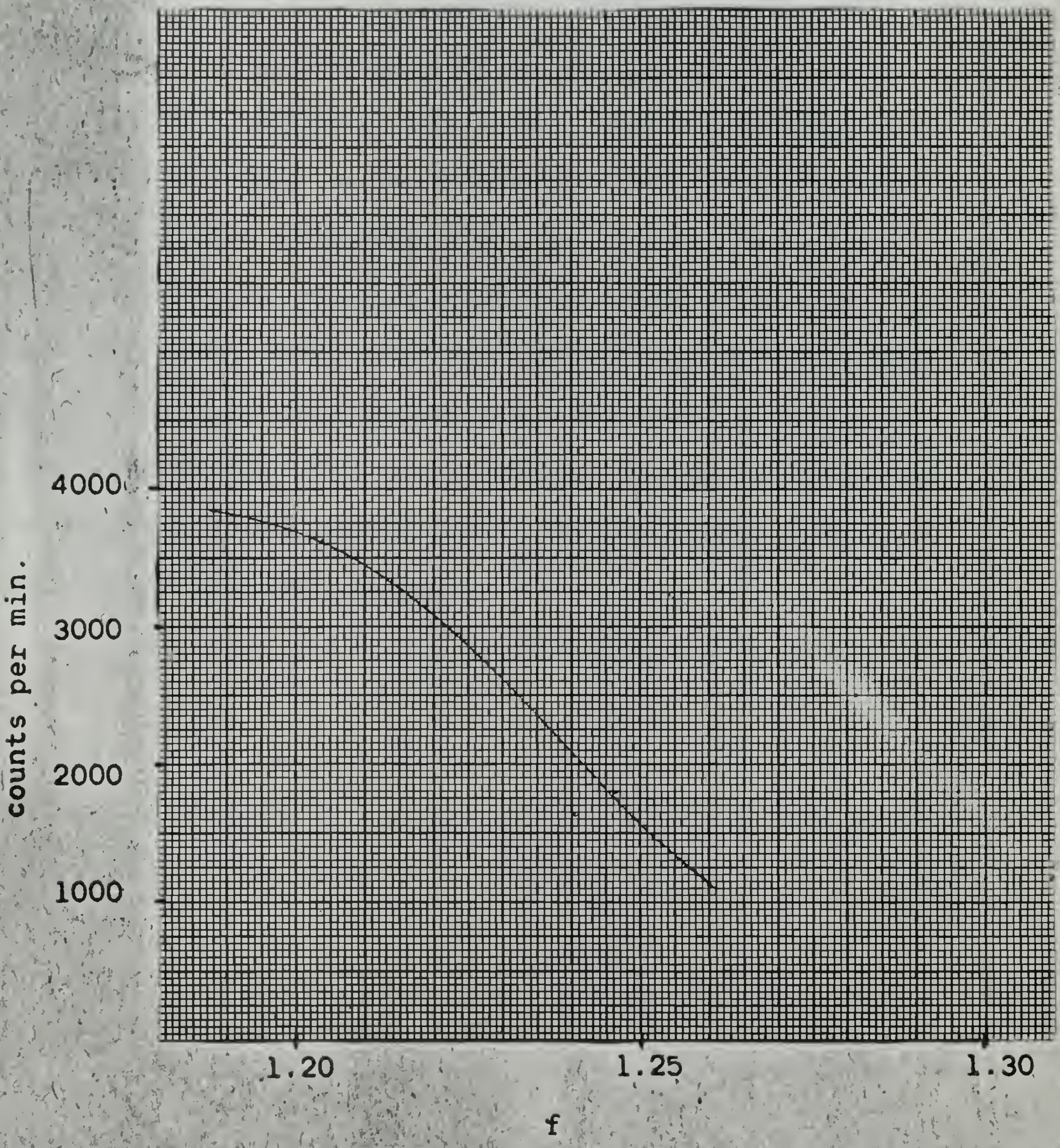

fig. V-7

This implied that the Cs peak was approximately at E_o. The curve of Figure V-7 could then be used to calibrate the E dial in energy. Application of this procedure before and after decay runs could be used to minimize and evaluate errors in energy calibration.

It was found that the counting rate of the differential pulse height spectrum near the peak of the standard Cs^{137} source had changed over a period of some weeks. This implies that either the source positioning or the differential pulse height analyzer calibration changed during that time. For this method to have good reliability, the complete calibration should be checked periodically. The isomers that were measured with the differential analyzer had very little contamination. Therefore, a small error in energy calibration would not be expected to cause a significant error in the half-life determination. This was borne out by the results of the decay measurements.

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